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Universal Upper Bound for the Tunneling Rate of a Large Quantum Spin

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Abstract. An upper bound is derived for the tunneling rate of a spin with large spin quantum number S. The bound is *universal* in the sense that it does not depend on the specific form of the anisotropy (i.e., the potential barrier). The method of proof relies on the exponential localization theorem of Fröhlich and Lieb and lends precise support to a rather suggestive interpretation put forth in a WKB analysis of van Hemmen and Sütő. The resulting bound agrees with their expression for the tunneling rate in the limit of large S.

1. Introduction

Macroscopic quantum tunneling, i.e., the penetration of a classically forbidden barrier by a macroscopic system, has aroused a considerable amount of interest [1]. The motion of the system, a collection of particles, is usually described by a single coordinate which is allowed to tunnel through a barrier between two minima of an effective potential. However, not only a system of particles but also a large quantum spin can tunnel [2–6]. For example, at low temperatures the long-time behavior of the thermoremanent magnetization (TRM) of a spin glass with uniaxial or unidirectional anisotropy is dominated [7] by the tunneling of large, mainly ferromagnetic clusters. The same type of dynamics also occurs in magnetically anisotropic media [8] where the magnetization of a single domain can tunnel through an energy barrier between easy directions. Since at low temperatures the clusters are frozen (i.e., the magnetic moments stick together), it is reasonable to describe them [7, 8], at least in first approximation, by a *single* spin with a *large* spin quantum number S.

We consider a single quantum spin of fixed total angular momentum S and denote by $\hat{S}_x$, $\hat{S}_y$, and $\hat{S}_z$ the usual angular momentum operators with

$$[\hat{S}_x, \hat{S}_y] = i\hbar \hat{S}_z, \quad \text{and cyclically; } \hat{S}_\pm = \hat{S}_x \pm i\hat{S}_y. \tag{1.1}$$

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Without loss of generality we can assume that the main anisotropy axis is the z-axis, choose a representation of (1.1) *with* $\hat{S}_z$ *diagonal*, and take $\hbar = 1$.

A large class of models is given by a Hamiltonian of the form [3, 4]

$$\mathscr{H} = -F(\hat{S}_z) - \frac{1}{2}\sum_{n=1}^{N} \alpha_n(\hat{S}_+^n + \hat{S}_-^n). \tag{1.2}$$

The first term on the right, $-F(\hat{S}_z)$, describes the anisotropy. The function F is *strictly* convex[1], even, and $F(0) = 0$; for instance, $F(s) = \gamma|s|^l$ with $l \geqq 1$ and $-S \leqq s \leqq S$ will do. The function $-F$ has two *degenerate* minima at $\pm S$. The second term in (1.2), a linear combination of powers of $\hat{S}_+$ and $\hat{S}_-$, tunnels the spin through the anisotropy barrier, which should be classically impenetrable. We, therefore, require

$$F(S) \gg \sum_{n=1}^{N} \alpha_n S^n. \tag{1.3}$$

Throughout what follows, S is an integer. (Half-integer S may give rise to Kramers degeneracy.)

Under the above conditions, van Hemmen and Sütő [3, 4] have derived a rather precise expression for the tunneling rate τ^{-1} by devising a novel but nonrigorous WKB argument. A surprising result is the *universality* [3, 4] of the tunneling rate. For low enough energies, say $E \approx -F(S)$, and in the limit of large S it is found that

$$\tau^{-1} = \tau_0^{-1}(\alpha_N S^N/|E|)^{2S/N}, \tag{1.4}$$

where

$$\tau_0^{-1} = F'(S)/2\pi \tag{1.5}$$

is an attempt frequency. Except for τ_0^{-1}, the tunneling rate is *independent* of the specific form of the anisotropy F and governed only by the energy E and the highest degree of the transverse field. The universality itself is similar to the one found in the particle case [9], but we will see shortly that the methods of proof do not bear any resemblance whatsoever.

A rather suggestive interpretation of (1.4) has been put forth in Ref. 3: "In tunneling the spin has to pass a barrier of height $|E|$. The driving force in the Hamiltonian is either $\hat{S}_+^N$ or $\hat{S}_-^N$ and the energy associated with each of them is $\alpha_N S^N$. This gives the fraction $x = \alpha_N S^N/|E| \ll 1$. Since $E \lesssim -F(S)$, the spin has to travel a distance $2S$ (in units $\hbar$) and the operators $\hat{S}_\pm^N$ force it to do N steps a time. After $k = 2S/N$ times the spin has reached the other side. Thus $\tau^{-1} = \tau_0^{-1} x^k$, where τ_0^{-1} is the attempt frequency." The authors of ref. 3 then continued by saying "Of course, the argument should not be taken too literally."

In this paper we derive an exact upper bound for the tunneling rate, which in the limit of large S reduces to (1.4). The method of proof relies on the exponential localization theorem of Fröhlich and Lieb [10] and lends mathematically precise support to the above interpretation. As a preparation we derive in Sect. 2 an

[1] In the WKB analysis of ref. 4 only convexity "at large distances" is required. The *strict* convexity is a technical requirement which we need in Sect. 3

expression for the level splitting ΔE by exploiting a symmetry of the Hamiltonian. Section 3 is devoted to the exponential localization work and Sect. 4 is a discussion.

2. Symmetry and Level Splitting

Hamiltonians which allow tunneling usually have a symmetry. In our case the Hamiltonian (1.2) is invariant under a rotation $\mathcal{R}$ through π about the x-axis. This rotation transforms $\hat{S}_z$ into $-\hat{S}_z$ and leaves the tunneling term invariant. Taking advantage of the symmetry $\mathcal{R}$ we derive a formula for the level splitting ΔE associated with tunneling. In ref. 4, Appendix C, it has been shown that for the low-lying states the tunneling rate τ^{-1} equals $|\Delta E|/\pi\hbar$; here $\hbar = 1$. So it suffices to compute ΔE.

For the sake of clarity we start by considering

$$\mathcal{H} = -F(\hat{S}_z) - \alpha\hat{S}_x, \tag{2.1}$$

and indicate later on how the arguments can be generalized so as to cover (1.2) with arbitrary N. The term $\alpha\hat{S}_x$ is supposed to be "small" compared to $-F(\hat{S}_z)$, i.e., $\alpha S \ll F(S)$; cf. (1.3). For $\alpha = 0$, $\mathcal{H} = -F(\hat{S}_z)$ has two-fold degenerate eigenvalues $-F(m)$, $1 \leq m \leq S$, and a simple eigenvalue at zero, which will be discarded throughout what follows. A simple perturbation calculus [11] shows that the degeneracy is lifted for $\alpha \neq 0$ (and S integer). By (1.3), the level splitting ΔE is quite small, in particular for the low-lying states.

The rotation $\mathcal{R}$ which leaves the Hamiltonian (2.1) invariant is unitarily implemented by a transformation which we again call $\mathcal{R}$. For even S we have $(\mathcal{R}\phi)(m) = \phi(-m)$, whereas for odd S we obtain $(\mathcal{R}\phi)(m) = -\phi(-m)$; cf. ref. 12. (Note that we work in the spectral representation of $\hat{S}_z$.) If ϕ is an eigenfunction of $\mathcal{H}$, then so is $\mathcal{R}\phi$ and, therefore, so are $(\phi \pm \mathcal{R}\phi)$. Since for $\alpha \neq 0$ the eigenspaces of $\mathcal{H}$ are one-dimensional, the corresponding eigenfunctions must be either even or odd. We now show that they always occur in pairs of *opposite* parity.

As $\alpha \to 0$, we can order the eigenfunctions in pairs $\{\phi_{2m}, \phi_{2m+1}\}$ whose linear span converges to the eigenspace of $-F(\hat{S}_z)$ which corresponds to the eigenvalue $-F(m)$ and is spanned by ψ_m and ψ_{-m} with $\psi_{\pm m}(n) = \delta_{n,\pm m}$. In the same limit one finds [11]

$$\left\| \phi - \frac{1}{\sqrt{2}}(\psi_m \pm \psi_{-m}) \right\| \to 0, \tag{2.2}$$

where ϕ stands for either ϕ_{2m} or ϕ_{2m+1} and $\|\cdot\| = (\cdot,\cdot)^{1/2}$ is the usual Euclidean norm. The symmetry $\mathcal{R}$ and the orthonormality dictate the coefficients of ψ_m and ψ_{-m} in (2.2). Hence, by continuity in α, ϕ_{2m} has to be even and ϕ_{2m+1} has to be odd, or the other way around. Which of them is even depends on the Hamiltonian. Let us call the even on ϕ_+, the odd one ϕ_-, put $\mathcal{H}\phi_\pm = E_\pm \phi_\pm$, and define $\Delta E = E_+ - E_-$.

We now turn to the computation of ΔE. The following definition was inspired by a beautiful paper of Harrell's [13]. Let

$$(\hat{I}\phi)(m) = \operatorname{sgn}(m)\phi(m), \quad -S \leq m \leq S, \tag{2.3}$$

where sgn(m) is the sign of m, vanishing at $m=0$. The operator $\hat{I}$ is hermitian. By (2.2) we can assume

$$(\hat{I}\phi_-, \phi_+) = 1 + O(\alpha). \tag{2.4}$$

Because $(\hat{I}\phi_-, \mathscr{H}\phi_+) = (\hat{I}\phi_-, \phi_+)E_+$ and $(\mathscr{H}\phi_-, \hat{I}\phi_+) = E_-(\hat{I}\phi_-, \phi_+)$, we immediately get

$$\Delta E = E_+ - E_- = \frac{(\phi_-, [\hat{I}, \mathscr{H}]\phi_+)}{(\hat{I}\phi_-, \phi_+)}, \tag{2.5}$$

and all we have to do is calculate the commutator $[\hat{I}, \mathscr{H}]$. Plainly, $\hat{I}$ and $F(\hat{S}_z)$ commute. To compute $[\hat{I}, \hat{S}_x]$, we define

$$a(m) = [S(S+1) - m^2]^{1/2} \tag{2.6}$$

for $|m| \leqq S$ and $a(m) = 0$ for $|m| > S$, note that

$$(\hat{S}_\pm \phi)(m) = a(\sqrt{m(m \mp 1)})\phi(m \mp 1), \tag{2.7}$$

and find

$$\begin{aligned}([\hat{I}, \hat{S}_x]\phi)(m) = \tfrac{1}{2}\{&a(\sqrt{m(m-1)})[\mathrm{sgn}(m) - \mathrm{sgn}(m-1)]\phi(m-1) \\ &+ a(\sqrt{m(m+1)})[\mathrm{sgn}(m) - \mathrm{sgn}(m+1)]\phi(m+1)\}.\end{aligned} \tag{2.8}$$

The right-hand side of (2.8) vanishes for all m except for $|m| \leqq 1$, where

$$([\hat{I}, \hat{S}_x]\phi)(m) = \begin{cases} \tfrac{1}{2}a(0)\phi(0), & m = 1, \\ \tfrac{1}{2}a(0)[\phi(-1) - \phi(1)], & m = 0, \\ -\tfrac{1}{2}a(0)\phi(0), & m = -1. \end{cases} \tag{2.9}$$

In passing we note that, in view of (2.7) and the non-degeneracy of the eigenvalues, there is no harm in assuming the eigenfunctions $\phi_\pm$ to be real. This is consistent with (2.2).

Combining (2.5)–(2.9) we then arrive at a surprisingly simple result.

Proposition. *The level splitting ΔE associated with the Hamiltonian (2.1) is*

$$\Delta E = -\alpha a(0)\phi_+(0)\phi_-(1)[1 + O(\alpha)]. \tag{2.10}$$

In the case of the more general Hamiltonian (1.2) one gets instead of (2.10) a linear combination of products $\phi_+(p)\phi_-(q)$ with $0 \leqq p$, $q \leqq N$. Here N is the maximal range of the walk induced by $\hat{S}_\pm^n$, $1 \leqq n \leqq N$, on the spectrum of $\hat{S}_z$. As $S \to \infty$, the terms $\phi_+(p)\phi_-(q)$ refer to a fixed and finite region well beyond the classically allowed domain and, therefore, are expected to be exponentially small. The proof of this statement is the subject of the next section.

3. Exponential Localization

Some years ago Fröhlich and Lieb [10] proved quite a remarkable theorem, which they tagged "exponential localization of eigenvectors." The content of the theorem is the following. Given a Hilbert space, let A and B be selfadjoint operators (typically finite, hermitian matrices) such that (i) $A \geqq 0$, (ii) $\pm B \leqq \varepsilon A$ with $0 \leqq \varepsilon < 1$. Furthermore, let ϕ be an eigenvector of $A + B$ with $(A+B)\phi = \lambda\phi$ and $\|\phi\| = 1$. Choose some $\rho > \lambda \geqq 0$ such that

$$\sigma \equiv \varepsilon\rho(\rho - \lambda)^{-1} < 1. \tag{3.1}$$

Let M_ρ be the linear span of all the eigenvectors of A corresponding to eigenvalues $\geqq \rho$. (Plainly, $A - \lambda$ restricted to M_ρ is strictly positive and, thus, invertible). Finally, let $\psi \in M_\rho$ be a unit vector with the property

$$\text{(iii)} \qquad [B(A-\lambda)^{-1}]^j \psi \in M_\rho \tag{3.2}$$

for $0 \leqq j \leqq d-1$, with $d \geqq 1$. Then

$$|(\phi, \psi)| \leqq \sigma^d, \tag{3.3}$$

which is *exponentially small* in d.

We take[2]

$$A = [F(S) - F(\hat{S}_z)] + F(S) \quad \text{and} \quad B = -\alpha\hat{S}_x, \tag{3.4}$$

so that $A \geqq 0$ and $\pm B \leqq \varepsilon A$ if $\varepsilon = \alpha S/F(S)$. Since up to an additive constant $\mathscr{H}$ equals $A + B$ we can stick to (3.4) if we want to estimate $\phi_+(0)$ and $\phi_-(1)$ in (2.10). As we will see shortly, they are both of the same order of magnitude, so we concentrate on $\phi_+(0) = (\delta_0, \phi_+)$, where $\delta_0(m) = \delta_{m,0}$ is the eigenfunction of A that is ψ in (3.2).

We are interested in the level splitting associated with the ground state of $\mathscr{H} = A + B$, so $\lambda = F(S) + O(\alpha S)$ for either E_+ or E_-. Naively, it is expected that the "best" value for σ^d in (3.3) is obtained by taking d as large as possible, i.e., $d = S$. Then $\rho = [F(S) - F(S-1)] + F(S)$ and $\hat{S}_x = \frac{1}{2}(\hat{S}_+ + \hat{S}_-)$ can operate $(S-1)$ times on $\psi = \delta_0$ *before* leaving the space M_ρ which contains all the eigenfunctions of A with eigenvalues $\geqq \rho$. If $\rho(\rho-\lambda)^{-1}$ were 1 (it is not), then σ^d would reduce to ε^S and $\phi_+(0)\phi_-(1) \sim \varepsilon^{2S}$, as announced. In reality (see below), the parameters ρ and d are not independent and by varying d in the limit of large S we can *saturate* the bound given by (1.4). In this limit the attempt frequency τ_0^{-1} is subdominant and, therefore, will be discarded. Furthermore, (1.4) may be rewritten

$$\tau^{-1} \propto (\alpha S/|F(S)|)^{2S} \equiv \varepsilon^{2S}. \tag{3.5}$$

After these introductory remarks we now turn to a more precise statement.

Theorem. *For large S the tunneling rate associated with the ground state of the Hamiltonian (2.1) obeys the upper bound*

$$\tau^{-1} \leqq \alpha\pi^{-1} a(0) \exp\{2x[\ln\varepsilon + \Theta(x)]\}. \tag{3.6}$$

Here $x \leqq 2S$ depends on $\varepsilon = \alpha S/F(S)$ and either equals S (then $\Theta(x)$ vanishes) or approaches S as $\varepsilon \downarrow 0$. In this limit, $\Theta(x)$ becomes negligible as compared to $\ln\varepsilon$.

[2] Other ways of splitting $\mathscr{H}$ into $A + B$ will do as well

Remark. Within the present context the generalization which is needed to cope with (1.2) and $(\hat{S}_+^n + \hat{S}_-^n)$, $n > 1$, is rather straightforward and will not be given explicitly.

Proof. Let $1 \leqq x \leqq S$ and $\rho = 2F(S) - F(x-1)$, so that $d = x$ in (3.3). We choose x in such a way that σ^x with $\sigma = \varepsilon\rho(\rho - \lambda)^{-1}$ is minimal. For S fixed, we then show that x either equals S or approaches S as $\varepsilon \downarrow 0$.

One readily verifies that $\sigma^x = \Phi(x)$, where

$$\Phi(x) = \varepsilon^x \left[\frac{2F(S) - F(x-1)}{F(S) - F(x-1)} \right]^x . \tag{3.7}$$

As F is strictly convex we have dropped $O(\alpha S)$ from the denominator in (3.7). The function $\ln \Phi$ and, hence, Φ itself is easily shown to be *convex* for $1 \leqq x \leqq S$. The derivative of $\ln \Phi$ at $x = 1$ is $\ln(2\varepsilon)$, which is negative by assumption ($\varepsilon \ll 1$). At $x = S$ we either have a negative derivative (in which case, e.g. for $F(x) = \exp(x^2) - 1$, we are done) or the derivative is positive, as is usually the case in practical work. Take, for instance, $F(x) = \gamma |x|^l$ for some $l > 1$. Then there exists a *unique* minimum for $1 < x < S$ and $x = x(\varepsilon)$ satisfies the equation

$$-\ln \varepsilon - \ln \left[1 + \frac{F(S)}{F(S) - F(x-1)} \right] = \frac{xF'(x-1)F(S)}{(2F(S) - F(x-1))(F(S) - F(x-1))} . \tag{3.8}$$

The left-hand side of (3.8) equals $-\ln \varepsilon - \Theta(x)$. This defines $\Theta(x)$ with $x = x(\varepsilon)$ in (3.6). The right-hand side of (3.8) is a monotonically increasing function of x. As $\varepsilon \downarrow 0$, $x = x(\varepsilon)$ has to increase to S, though the dependence upon ε is weak and, therefore, $\Theta(x)$ is subdominant as compared to $\ln \varepsilon$. By (3.3) and (2.10) the proof is finished.

4. Discussion

Rather surprisingly, the arguments of Sect. 3 do not make any reference whatsoever to the tunneling process itself. This is the more remarkable since the WKB analysis [3, 4], which hinges on the very notion of tunneling, gives rise to essentially the same estimates for the tunneling rate τ^{-1} as $S \to \infty$ (or $\alpha \to 0$). The underlying philosophy, however, is fully confirmed by the exponential localization theorem of Fröhlich and Lieb [10], which provides us with an upper bound for τ^{-1} in terms of the range of the walk induced by $\hat{S}_\pm^N$ on the spectrum of $\hat{S}_z$. For the tunneling rate associated with the groundstate of the Hamiltonian (1.2) the message of Eqs. (2.10) and (3.3)–(3.6) is that starting in the center ($m \approx 0$) the $\hat{S}_\pm^N$ have reached the borders of the spectrum of $\hat{S}_z$ after S/N steps, whence $\tau^{-1} \propto \varepsilon^{2S/N}$ with $\varepsilon = \alpha_N S^N / |E|$—as advertized.

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References

1. Leggett, A. J., Chakravarty, S., Dorsey, A. T., Fisher, M. P. A., Garg, A., Zwerger, W.: Rev. Mod. Phys. **59**, 1 (1987)
2. Vourdas, A., Bishop, R. F.: J. Phys. **G11**, 95 (1985)
3. van Hemmen, J. L., Sütő, A.: Europhys. Lett. **1**, 481 (1986)
4. van Hemmen, J. L., Sütő, A.: Physica **141B**, 37 (1986)
5. Enz, M., Schilling, R.: J. Phys. **C19**, 1765 (1986)
6. Scharf, G., Wreszinski, W. F., van Hemmen, J. L.: J. Phys. **A20**, 4309 (1987)
7. van Hemmen, J. L., Sütő, A.: Z. Phys. **B61**, 263 (1985)
8. Chudnowsky, E. M., Gunther, L.: Quantum tunneling of magnetization in small ferromagnetic particles, Phys. Rev. Lett. **60**, 661 (1988)
9. Kirsch, W., Simon, B.: Universal lower bounds on eigenvalue splittings for one-dimensional Schrödinger operators. Commun. Math. Phys. **97**, 453 (1985), and references therein
10. Fröhlich, J., Lieb, E. H.: Phase transitions in anisotropic lattice spin systems. Commun. Math. Phys. **60**, 233 (1978)
11. Kato, T.: Perturbation theory for linear operators. Berlin, Heidelberg, New York: Springer Verlag 1966, Chap II
12. Brink, D. M., Satchler, G. R.: Angular momentum, 2nd ed. Oxford: Oxford University Press 1968, Eqs. (2.15), (2.17), and (2.18)
13. Harrell, E. M.: Double wells. Commun. Math. Phys. **75**, 239 (1980)

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